PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

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18.MAR2004*126403

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing *(day/month/year)*	16.03.2004
Applicant's or agent's file reference AJC/P053051WO	**IMPORTANT NOTIFICATION**

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB 03/00792	24.02.2003	28.02.2002

Applicant
BRITISH NUCLEAR FUELS PLC et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

28/8/04
9/4/04

Name and mailing address of the international preliminary examining authority:	Authorized Officer
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PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

REC'D 17 MAR 2004
WIPO PCT

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
AJC/P053051WO		
International application No. PCT/GB 03/00792	International filing date *(day/month/year)* 24.02.2003	Priority date *(day/month/year)* 28.02.2002
International Patent Classification (IPC) or both national classification and IPC G21C19/42		
Applicant BRITISH NUCLEAR FUELS PLC et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 6 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:

   I ☒ Basis of the opinion
   II ☐ Priority
   III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   IV ☐ Lack of unity of invention
   V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   VI ☐ Certain documents cited
   VII ☐ Certain defects in the international application
   VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
25.09.2003	16.03.2004
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Lohberger, S Telephone No. +49 89 2399-6723

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB 03/00792

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, Pages

1, 5-7	as originally filed
2, 3, 4, 4a	received on 30.01.2004 with letter of 30.01.2004

Claims, Numbers

1-25	received on 30.01.2004 with letter of 30.01.2004

Drawings, Sheets

1/1	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: . , which is:

   ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).

   ☐ the language of publication of the international application (under Rule 48.3(b)).

   ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   ☐ contained in the international application in written form.

   ☐ filed together with the international application in computer readable form.

   ☐ furnished subsequently to this Authority in written form.

   ☐ furnished subsequently to this Authority in computer readable form.

   ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   ☐ the description, pages:

   ☐ the claims, Nos.:

   ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-25
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-25
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-25
	No:	Claims	

2. Citations and explanations

   see separate sheet

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Reference is made to the following documents:

   D1: WO-A-0141152
   D2: US-A-5378325
   D3: WO-A-0040782
   D4: US-A-5650053
   D5: US-A-5160367
   D6: US-A-3982928
   D7: US-B1-6540902

2. Independent claim 1 now relates to an apparatus for performing a process for reducing to metallic form metal oxides **present in spent nuclear fuel**, wherein the apparatus comprises an electrochemical cell, which comprises a body or housing, a cathode container and a cathode connector, wherein the housing is maintained as the cathode and said electrochemical cell is free from bolted or screwed fittings.

   Claim 1 now is clear under article 6 PCT.

   Claim 13 relates to a process for reducing to metallic form metal oxides **present in spent nuclear fuel**, wherein the oxide is cathodically electrolysed in the presence of a molten salt electrolyte and the potential of the cathode is controlled so as to favour oxygen ionisation over deposition of metal from the cations present in the molten salt. The apparatus of claims 1 to 12 is used for this process.

3. Claim 1 is now in accordance with article 33(2) PCT.
   D2, especially description column 6, lines 49 to 60, figure 3 and claims discloses a

similar apparatus. Metal oxide is reduced by electrolysis into metal. A cathode container is used. Electrical current is transferred to this container by a cathode connector. No bolts or fittings can be found in the electrolysis cell. The body is maintained as the cathode. However this apparatus is not of the field of spent nuclear fuel with its specific constraints with regard to radiation protection and consequently such apparatus will not be used for treating radioactive material.

5. D1, especially description page 1, line 1 to page 4, line 26, figures and claims discloses some features of present claims 1 to 26. It is not stated that the apparatus should be free of bolted or screwed fittings only standard connecting means between cathode and circuit are mentioned.

   Consequently claims 1 to 26 are novel over D2 under article 33(2) PCT.

6. D4 discloses an anode basket and a cathode cylinder for electrochemical production of uranium. A bolted construction is used. D4 is not considered to be of relevance.

7. D5 relates to the chemical reduction of nuclear oxide fuels and is silent about any electrolytic reduction of nuclear oxide fuel or metal oxide.

8. D6 teaches the reduction of UO_2 by a pyrometallurgical process in molten salt. Electrolysis is not addressed.

9. D7 **which was published on Apr. 1, 2003**, discloses on column 2, line 5 to column 7, line 35, figures and claims the direct electrolytic reduction of metal oxide to the corresponding metal. The apparatus is free from bolted or screwed fittings. Formally D7 attacks novelty of claims 1 to 26 if present priority is not valid.

10. D3 does not relate to the field of radioactive spent nuclear fuel.

11. The description is properly adapted to the present set of claims. D2, D3 and D7 are cited in the introductory part of the description and are briefly discussed.

12. Present claim 1 (apparatus) and present claim 13 (process using this apparatus) now are based on an inventive step over the cited prior art in that the absence of bolted and screwed fittings in the electrolytic cell facilitates the reducing process in that no removal of bolted or screwed fittings from the apparatus is needed for the production of the metal from spent nuclear fuel metal oxide.

Preferably the oxide is in contact with the cathode container and it is preferred that the cathode is in the form of a mesh basket or, most preferably, an assembly of mesh baskets, with the oxide being contained within the said baskets. In this case, contact between the assembly of cathode containers and the cathode connectors is most simply achieved when the cathode connectors are in the form of a multiplicity of cathode rails which are welded to the base of the cell, allowing press contact to be brought about by the weight of the oxide feedstock in the cathode basket. This represents the most preferred embodiment of the first aspect of the invention. The anode may be any suitable inert anode, such as carbon.

According to a second aspect of the present invention, there is provided a process for reducing to metallic form metal oxides, the process comprising cathodically electrolysing the oxide in the presence of a molten salt electrolyte in an apparatus according to the first aspect of the invention, the potential of the cathode being controlled so as to favour oxygen ionisation over deposition of metal from the cations present in the molten salt.

The molten salt electrolyte may be any suitable molten salt or mixture of such salts, for instance chloride salts, preferably $CaCl_2$ and/or $BaCl_2$.

Preferably, the oxide treated by the process according to the second aspect of the invention comprises an oxide present in spent nuclear fuel. Typically, the oxide comprises an actinide oxide, such as uranium oxide or irradiated uranium oxide, or mixed uranium/plutonium oxides. The uranium oxide is commonly uranium dioxide. Alternatively, the oxide may comprise the oxide of a metal such as zirconium or hafnium.

In such a process, wherein the oxide comprises an oxide present in spent nuclear fuel, the fuel may be first treated mechanically to remove its zircaloy cladding before it is added to the electrolytic cell. Alternatively, the zircaloy cladding may be treated with

Statements of Invention

Thus, according to a first aspect of the present invention there is provided an apparatus for performing a process for reducing to metallic form metal oxides, the said apparatus being free from bolted or screwed fittings, and comprising an electrochemical cell which comprises a body or housing, a cathode container, and a cathode connector, wherein said body or housing is maintained as the cathode.

The body or housing of the cell which comprises the apparatus according to the first aspect of the invention is most conveniently maintained as the cathode by the provision of an electrical connection from a power supply, provided by means of a connector from the cathode terminal to the body or housing of the cell. Typically, said connector comprises a bolted connection, but this is positioned externally to the cell. No bolted or screwed connections are present within the cell, wherein a cathode connector is provided which is affixed to an internal surface of the cell, most preferably the base of the cell, generally by welding. Preferably said cathode connector comprises a cathode rail.

In operation, the body or housing of the cell is maintained as the cathode, and said cathode is brought into contact with the cathode container by means of the cathode connector. Thus, contact is made between the cathode container and the cathode connector in order to facilitate the electrolytic process. Contact may be most conveniently achieved by means of a simple press connection between the two components.

The cathode container preferably comprises a basket, such as a mesh basket, or vessel, typically a metal oxide retaining vessel, and – most preferably – comprises an assembly of such baskets or vessels. In order to effect electrical connection between such an assembly of cathode containers and the cell body during operation of the cell, it is necessary to provide a multiplicity of connectors and to effect contact between individual cathode baskets or vessels and individual connectors, preferably by means of a multiplicity of press connections.

molten salt, or a Li/Cd alloy, at 500 to 600°C. Alternatively, a salt transport process can be used involving a Cu-Mg-Ca alloy and molten $CaCl_2$ salt. However, in both reduction methods the by-products, Li_2O and CaO respectively, need to be recovered from the molten salt phase by an electrolysis step. Effectively this means a two stage process.

A disadvantage of the lithium reduction process for producing a metallic feed from an oxide is the production of Li_2O by-product. This requires recycle to make the process economic, and this is done by an electrolytic recovery of lithium metal. Hence this is a two stage process, comprising a reduction step followed by a lithium recovery stage.

In co-pending PCT patent application WO 01/41152 there is disclosed a single step process for reducing to metallic form a metal oxide present in spent nuclear fuel, the process comprising cathodically electrolysing the oxide in the presence of a molten salt electrolyte, the potential of the cathode being controlled so as to favour oxygen ionisation over deposition of the metal from the cations present in the molten salt.

The process thereby involves the use of a single electrochemical process to reduce the metal oxide fuel to a metallic form, with oxygen, CO or CO_2 produced as the only by-products. The potential of the cathode is maintained and controlled so that only oxygen ionisation occurs and not the deposition of metal (eg Ca) from the cations (eg Ca ions) in the fused salt. Typically, the oxide comprises an actinide oxide, such as uranium oxide or irradiated uranium oxide.

The present inventors have, however, now effected an improvement to the process described in WO 01/41152 which allows a more practical, efficient and financially viable process to be adopted in the production of metals from oxides. The new process is particularly beneficial in that it facilitates the removal of bolted and screwed fittings from the apparatus used for the production of the metal.

CLAIMS

1. An apparatus for performing a process for reducing to metallic form metal oxides, the said apparatus being free from bolted or screwed fittings and comprising an electrochemical cell which comprises a body or housing, a cathode container, and a cathode connector, wherein said body or housing is maintained as the cathode.

2. An apparatus as claimed in claim 1 wherein said cathode connector is affixed to an internal surface of the cell.

3. An apparatus as claimed in claim 2 wherein said cathode connector is affixed to an internal surface of the cell by means of welding.

4. An apparatus as claimed in any one of claims 1, 2 or 3 wherein an electrical connection from the cathode container to the body or housing of the cell is provided by means of a cathode connector.

5. An apparatus as claimed in claim 4 wherein said electrical connection is provided by means of a press connection.

6. An apparatus as claimed in any preceding claim wherein said cathode connector comprises a cathode rail.

7. An apparatus as claimed in claim 6 wherein said rail is welded to the base of the cell.

8. An apparatus as claimed in any preceding claim wherein the cathode container comprises a mesh basket or metal oxide retaining vessel.

9. An apparatus as claimed in any preceding claim wherein the cathode container comprises an assembly of cathode containers.

10. An apparatus as claimed in claim 9 wherein said assembly comprises an assembly of mesh baskets or metal oxide retaining vessels.

11. An apparatus as claimed in any preceding claim wherein the anode is a carbon anode.

12. An apparatus as claimed in any preceding claim wherein the body or housing of the cell is maintained as the cathode by the provision of an electrical connection from a power supply to the body or housing of the cell.

13. A process for reducing to metallic form metal oxides, the process comprising cathodically electrolysing the oxide in the presence of a molten salt electrolyte in an apparatus as claimed in any one of claims 1 to 12, the potential of the cathode being controlled so as to favour oxygen ionisation over deposition of metal from the cations present in the molten salt.

14. A process as claimed in claim 13 wherein the body or housing of the cell is maintained as the cathode and is brought into contact with the cathode container by means of a press connection between said container and a cathode connector.

15. A process as claimed in claim 13 or 14 wherein the oxide comprises the oxide of zirconium or hafnium.

16. A process as claimed in claim 13 or 14 wherein the oxide comprises an oxide present in spent nuclear fuel.

17. A process as claimed in claim 16 wherein the oxide comprises an actinide oxide.

18. A process as claimed in claim 17 wherein the actinide oxide comprises uranium oxide, irradiated uranium oxide or mixed uranium/plutonium oxide fuel pellets.

19. A process as claimed in claim 18 wherein the uranium oxide comprises uranium dioxide.

20. A process as claimed in any of claims 13 to 19 wherein the oxide is located in a mesh basket which forms the cathode.

21. A process as claimed in any of claims 13 to 20 wherein the molten salt electrolyte comprises at least one chloride salt.

22. A process as claimed in claim 21 wherein the chloride salt is $CaCl_2$ or $BaCl_2$.

23. A process as claimed in any one of claims 16 to 22 wherein the fuel is treated together with its cladding.

24. A process as claimed in any one of claims 16 to 22 wherein the cladding is removed from the fuel prior to treatment.

25. A process as claimed in any of claims 13 to 24 wherein the metal resulting from the process is used as the feed for an electrorefining process.

26. A process as claimed in claim 25 wherein the electrorefining process is carried out in the same electrolytic cell as the electrolytic reduction process.